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# Composition and mixing states of brown haze particle over the Himalayas along two transboundary south-north transects

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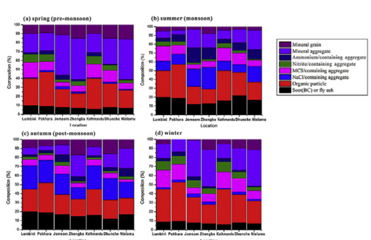
## HIGHLIGHTS

- We present the composition and mixing states of brown haze particle over the Himalayas.
- BC-fly ash, dust, salt and organic particles were often internally mixed together.
- Large proportion of individual particles (25–56%) appeared as salt-coated particles.
- Obvious spatial and seasonal change of various components were observed.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Pollutants that are usually transported from southern Asia to the Tibetan Plateau deposit on the Plateau surface, change snow albedo and thereby surface radiative flux. This results numerous climatic implications like as erratic monsoon, perturbation in hydrological cycle, etc. However, the accurate estimation of these climatic implications is not well understood, because the atmospheric pollution is a heterogeneous mixture of various particle types. Therefore, this part of climate research requires a detailed investigation of physical and chemical properties of atmospheric pollutants. This study aimed to examine the physical and chemical properties of atmospheric pollutants across the Himalayan regions along two transboundary south-north transects. The information of individual-particles was obtained using microscopy-based techniques that comprises transmission electron microscope (TEM) and Energy-dispersive X-ray spectrometer (EDX). Study capture the signatures of various types of atmospheric species such as black carbon (BC), mineral dust, fly ash, organic matter, sulfate, nitrite, ammonium, and NaCl. Microscopy-based techniques confirm that these particles were generally in mixing state, for example salt-coated particles accounting for 25–56% of the total particles in sampled locations. Our analysis shows that urban and rural locations are characterized with atmospheric particles which sourced from anthropogenic activities, whereas remote locations with those released from natural crustal. However, the relative contributions of anthropogenic particles were higher than that of particles

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released from natural crustal. The presence of such particles over remote locations of Himalayan region provides an evidence of prevailing atmospheric transport processes, which further need to be well understood. It is expected that this work would be helpful in understanding the regional atmospheric conditions and the transboundary transport process of haze particles. As these informations are of great importance in modeling studies, which further lead to improve understanding of haze particles climate effects.



## 1. Introduction

The Tibetan Plateau is one of the largest plateaus in the world having an area of ~2500000 km$^2$, hold largest ice mass outside the Polar Regions (Xu et al., 2009). These unique features are giving its name “The Third Pole” (Qiu, 2008). Its glaciers are the major source of rivers for example, Indus, Ganges, Brahmaputra, Yangtze, and Yellow Rivers (Immerzeel et al., 2010). These glacier fed rivers are the lifelines of millions of people living in the downstream (Singh and Bengtsson, 2004; Qiu, 2008; Xu et al., 2009). Tibetan Plateau plays a crucial role in regulating Asian monsoon (Singh and Bengtsson, 2004; Qiu, 2008; Xu et al., 2009). The rapid pace of urbanization and industrialization along the elevated site of the Himalaya have subsequently increased the burden of atmospheric pollution (Lau et al., 2006, 2008; Gautam et al., 2009; Guleria et al., 2011a, 2011b; Guleria and Kuniyal, 2016), which have adversely affected the Himalayan glaciers and hence the climate system (Ramanathan at el., 2001, 2007a; Lau et al., 2006, 2008). The high burden of particles particularly light absorbing can influence the snow albedo, energy balance of the atmosphere, and water exchange between the land surfaces and the lower troposphere (Takeuchi et al., 2001; Flanner et al., 2007; Anesio et al., 2009; Kaspari et al., 2011; Cong et al., 2015; Chen et al., 2015; Ji et al., 2015; Dong et al., 2016a, 2016b). The deposition of atmospheric pollution on Tibetan glaciers is an important factor responsible for rapid glacier retreat and thermal heating (Xu et al., 2009; Kang et al., 2010, 2016). The rate of warming in the Tibetan region is significantly higher than the global average (Qian et al., 2011). Therefore, climate change has become a major concern in the Tibetan regions and is a key issue of climate science research.

The Tibetan Plateau is a mid-latitudinal location, located in the vicinity of densely populated and industrialized regions (McConnell et al., 2007; Ming et al., 2013; Dong et al., 2014, 2015; Kang et al., 2015; Li et al., 2016a). There are hardly any anthropogenic activities on the Plateau surface (Dong et al., 2015; Qiu, 2008), inspite of that this region encounter with high atmospheric pollution that usually transport from the southern Asian countries (Xu et al., 2009; Li et al., 2016a, Fig. 1). South Asian countries are significant source of pollution such as organic matter and polycyclic aromatic hydrocarbons (Li et al., 2016b), causing severe regional environmental pollution on the Plateau (Lüthi et al., 2015). Earlier studies have identified the accumulation of atmospheric air pollution loading along the elevated sites particularly Indian Himalaya and its climate effects in the Himalayan regions (Lau et al., 2010; Gautam et al., 2011, 2013; Guleria et al., 2013a; 2013b, 2014). Fig. 2 showing the Cloud-Aerosol Lidar with Orthogonal Polarisation (CALIOP) vertical profile of aerosols over the Tibetan Plateau and southern Asia region and Fig. 3 showing air mass backward trajectory to the Tibetan locations (Nielamu and Zhongba), also captured the signatures of atmospheric pollutants transport from southern Asian countries (Figs. 2 and 3). Therefore, the Tibetan Plateau is considered one of the regions most vulnerable to effects of various types of aerosols. The long-range transport to and deposition of BC and salt (e.g. Nitrite and Sulfate) aerosols on the Tibetan Plateau is attracting considerable attention because of their effects on the transformation of hydrological and radiative forcing in the East and South Asian regions (Ramanathan et al., 2007a, 2007b, 2008; Menon et al., 2010). Moreover, BC aerosols are believed to play an important role in the glaciers melting because of both the heating of aloft air masses transported into the Tibetan Plateau and the albedo effects of deposited BC (Flanner et al., 2007; McConnell et al., 2007). During the past 50 years, Tibetan Plateau surface temperature showed a larger increase than that of the world average, which may also increase its environmental vulnerability (IPCC, 2013). Previous studies have reported the composition and characteristics of regional black carbon and organic pollutants in the region's atmosphere (Ramanathan et al., 2007b; Li et al., 2016a). However, previous work focused on only one or two kinds of special species of haze particles (e.g., BC and organic), and study remains very limited on the composition and mixing states of individual particles of various species in the brown haze of the southern Asian and Himalayan regions.

Atmospheric pollution, here also referred as light absorbing impurities (LAIs) has a different radiative forcing (RF) in the atmosphere and in glacier cryoconite (IPCC, 2013; Xu et al., 2009; Oerlemans et al., 2009; Wientjes et al., 2011; Qu et al., 2014; Naegeli et al., 2015; Dong et al., 2016a). For example the impurities of BC-soot particles in the ice core of Mt. Everest have produced a summer darkening effect of 4.5 W m$^{-2}$ (Ming et al., 2008). Thus, it is important to study the LAIs' particle components in the atmosphere. Moreover, transmission electron microscope (TEM) is a good method for learning the particle's morphology and physicochemical composition (Semeniuk et al., 2014; Dong et al., 2016a). For example, a study based on observation and TEM analysis has found that dust and biological aerosols from the Sahara and Asia may influence precipitation in the Western United States (Creamean et al., 2013). However, few studies have focused on the total suspended particles (TSP's) morphology, chemical composition and physical state in the trans-southern Asian-Himalayan-Tibetan regions.

The main objectives of this study are to provide a unique record of the particle's physical and chemical composition, mixing states of brown haze particle over the Himalayas, to determine its source and transport route under the influence of regional atmospheric circulation, to help understanding the regional atmospheric conditions and the transboundary transport process of haze particle and its climate effects. Moreover, till date our understanding on individual haze particle is very limited, because atmospheric processes are too complex and couldn't represent accurately in global climate models. Therefore, this work will finally help in improving atmospheric processes analysis in global climate models.

We organized the paper as follows: In section 2, we provided detailed descriptions about data and method of individual haze particle analysis; and in section 3 we presented the observed results and discussion of: (i) haze particle types and their mixing states in the atmosphere along two transects; (ii) spatial and